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UNIV BATON ROUGE DEPT OF PHYSICS AND ASTRONOMY
J CALLAWAY JAN 85 ARO-17235.7-PH DAAG29-81-K-0006

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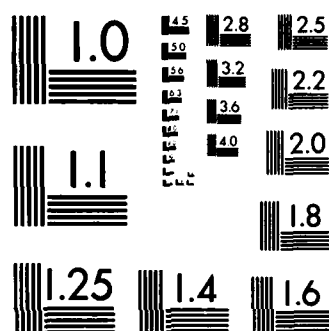
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20. ABSTRACT CONTINUED:

orbitals. Our intention was to use the clusters to study the effects of small concentrations of foreign atoms in transition metal hosts (example: carbon in iron), because calculations for clusters could be made self consistent to an extent not normally possible for impurities in bulk solids. Subsequently, experimental observations of the properties of iron clusters by research groups at the Exxon laboratories and elsewhere have stimulated us to consider free clusters in more detail.



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Final Report

Clusters of Transition Metal Atoms

Contract DAAG29-81-K-0006

Oct. 20, 1980 - December 19, 1984

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Statement of Problem

The problem addressed in the work supported was to develop a method for accurate all electron self consistent calculations of the electronic structure of small clusters of transition metal atoms on the basis of local spin density functional theory and using a basis of symmetrized combinations of Gaussian orbitals. Our intention was to use the clusters to study the effects of small concentrations of foreign atoms in transition metal hosts (example: carbon in iron), because calculations for clusters could be made self consistent to an extent not normally possible for impurities in bulk solids. Subsequently, experimental observations of the properties of iron clusters by research groups at the Exxon laboratories and elsewhere have stimulated us to consider free clusters in more detail.



Results

We begin our work by attempting to modify the standard quantum chemistry program GAUSS76 to perform density functional calculations. This attempt was successful for small systems (diatomic molecules). The results obtained for LiH, CO and NO indicate that spin density functional theory leads to an excellent account of the ground state properties of simple molecules. However, it did not prove feasible to study the large (15 - 20 atom) systems of primary interest to us with this approach.

Instead we developed our own program. The essential features are that it makes extensive use of symmetry to enable efficient calculations for large systems. An auxiliary basis was introduced for variational fitting of the charge density to eliminate the necessity for the computation of four center-two electron integrals. A numerical grid was developed for the evaluation of matrix elements of the exchange correlation potential. We believe this program is the most accurate available at the present time for cluster calculations.

Results were obtained for the following clusters: Fe_9 , Fe_{15} , Fe_{14}C (with body centered cubic geometry) and Fe_{13} , Ni_{13} , and Ni_{19} (with face centered cubic geometry). The atomic spacing was taken to that of the bulk metal (the geometries of experimentally observed clusters are unknown). Specific results are described in detail in the publications which are listed subsequently. In a general way, they may be summarized as follows:

The general features of the density of state for the cluster resemble those of the bulk metal as obtained from band calculations for clusters as small as 15 atoms. This was particularly striking for the case of the Fe_{15} cluster: Not only does the overall width in energy of the d level complex in

the cluster agree with that of the bulk but the positions and heights of several of the major peaks in the density of states correspond as well. Our results for clusters are in much better agreement with those for the solid than are those of other workers who have used different methods (DVM, MS-X α), and support the view that the density of states depends only weakly on the boundary conditions imposed.

All of the clusters studied were ferromagnetic; i.e., have net magnetic moments. The moments are larger than for the bulk, roughly $3\mu_B$ /atom for the iron clusters. This has turned out to be in agreement with observation. However the spatial distribution of the spin density in the iron clusters is quite different from that of the bulk. The central atom has more minority (down) spin electrons than majority (up) spin. This is quite pronounced in the case of the f.c.c. Fe₁₃ cluster, where there is a minority spin excess on the central atom of about 2 electrons. In a sense, the Fe₁₃ cluster can be regarded as similar to an antiferromagnet. But in contrast, the nickel clusters studied do not exhibit this effect: both the value of the average moment and its spatial distribution are closer to that of the bulk metal than for iron.

These results are in accord with the ideas of spin density functional theory in regard to the tendency of the exchange correlation potential to induce a spatial separation of majority and minority spin densities. This tendency is much more pronounced in iron clusters than in nickel because the atomic moment is larger.

The ionization potentials of the Fe₉ and Fe₁₃ clusters have been determined by separate, self consistent, transition state calculations.

In an independent calculation of basic importance to density functional theory, we have calculated the temperature dependence of the exchange

correlation potential for a spin polarized electron gas over a wide range of temperatures. Relatively low temperatures are relevant to solid state physics, and in this situation we find that the leading (T^2) temperature dependence of the exchange and correlation potentials cancel: over the range of temperatures of interest to solid state physics, the temperature dependence of the exchange correlation potential is negligible except possibly in circumstances in which the electron density is exceptionally low.

Publications

1. A Linear Approximation in Self Consistent Density Functional Calculations for Atomic Clusters. J. Callaway, S. Ramasesha, and D. G. Kanhere, Phys. Letts. 94, 139(1983).
2. Electronic Structure of Small Iron Clusters. K. Lee, J. Callaway, and S. Dhar, Phys. Rev. B 30, 1724(1984).
3. Electronic Structure of Some Heteronuclear Diatomic Molecules in the Local Spin Density Approximation. S. Dhar, A. Ziegler, D. G. Kanhere, and J. Callaway, accepted for publication in J. Chem. Phys.
4. Exchange Correlation Potentials for Spin Polarized Systems at Finite Temperatures. D. G. Kanhere, P. V. Panat, A. K. Rajagopal, and J. Callaway, submitted to Phys. Rev. A.
5. Electronic Structure of Small Iron Clusters. K. Lee, Ph.D. Thesis, Louisiana State Universtiy 1984, available from University Microfilms International, Ann. Arbor, Michigan, 48106.

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